An Updated Bibliography (1845-1986) On Ozone, its Biological Effects and Technical Applications

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Canadian Technical Report of
Fisheries and Aquatic Sciences No. 1542

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AN UPDATED BIBLIOGRAPHY (1845-1986) ON OZONE, ITS BIOLOGICAL EFFECTS AND TECHNICAL APPLICATIONS

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ABSTRACT

Rosenthal, H., and J. S. Wilson. 1987. An updated bibliography (1845-1986) on ozone, its biological effects and technical applications. Can. Tech. Rep. Fish. Aquat. Sci. 1542: vii + 249 p.

Interest in ozone as reflected in scientific literature has grown at a staggering rate over the last decade. This bibliography is an attempt to collate references to many of these publications. The bibliography concentrates on aquatic aspects of ozone preparation and generation, ozone determination and measurement, ozone chemical reactions and physical-chemical properties, municipal and industrial applications, water treatment applications and biological effects, but also includes much material from other areas of interest, such as atmospheric and stratospheric ozone, ozone removal, costs of ozonation, review articles and books, and extensive patent citations for ozone equipment. Most references are provided with one or more cross-references to an abstracting service for a quick check of the article's abstract. The bibliography contains 4790 references and an extensive subject index. The subject index is divided into 13 main subject categories and approximately 200 sub-categories.

RÉSUMÉ

Rosenthal, H., et J. S. Wilson. 1987. An updated bibliography (1845-1986) on ozone, its biological effects and technical applications. Can. Tech. Rep. Fish. Aquat. Sci. 1542: vii + 249 p.

L'intérêt suscité par l'ozone s'est accru à une allure vertigineuse au cours de la dernière décennie si l'on en juge par la place qu'occupe ce sujet dans la documentation scientifique. Dans la présente bibliographie, on a cherché à rassembler les titres d'un grand nombre de ces publications. La bibliographie porte surtout sur les aspects aquatiques de la préparation et de la production d'ozone, la caractérisation et de dosage de l'ozone, ses réactions chimiques, ses propriétés physico-chimiques, ainsi que ses applications municipales et industrielles, ses applications à l'épuration de l'eau et ses effets biologiques, mais elle cite également de nombreux documents portant sur d'autres questions d'intérêt, comme l'ozone atmosphérique et stratosphérique, la disparition de l'ozone et le côut de l'ozonisation, ainsi que des articles et des ouvrages de synthèse et de nombreux extraits de brevets relatifs à du matériel destiné à la production et à l'utilisation d'ozone. La plupart des références sont accompagneés d'au moins un renvoi à un service de résumés analytiques, ce qui permet de consulter rapidement les résumés. La bibliographie renferme 4790 références et un index exhaustif des sujets. Cet index est divisé en 13 catégories de sujets principaux et en 200 sous-catégories environ.

ACKNOWLEDGEMENTS

A project of this magnitude would not have been readily accomplished without considerable assistance and advice from friends and colleagues. In Germany where the project started, we must thank Elisabeth Meyer, Günter Krüner, Andrew Forsythe, and Rolf Berte of the Biologische Anstalt Helgoland for help with the computer set-up and for help with data entry. In Canada, the following people all assisted in collecting the references: Gary Batt and Maureen McInerney Northcott of the Atlantic Regional Laboratory, National Research Council of Canada (ARL, NRC), and Ingrid Rosenthal, Karin Rosenthal, Birgit Rosenthal, Madelyn Wilson, Patricia Miller, and Barbara Hoisington. The libraries and librarians of the ARL, NRC, the Department of Fisheries and Oceans (DFO) Halifax laboratory, the Bedford Institute of Oceanography, and Dalhousie University were all of great help in this project. Dr. James Craigie of ARL, NRC loaned us an Apple IIc computer which was used for most of the data entry. For final editing and printing, computer facilities at DFO were used. We must especially thank Jerry Black of DFO's Fisheries Research Branch for his guidance and helpfulness in data transfer and formatting in the Macintosh system. Christine Hunter and Dave Swetnam of DFO's Invertebrates and Marine Plants Division also willingly gave their help. Dr. Walter J. Blogoslawski kindly reviewed the introductory text. The project would not have been possible without the interest, encouragement and assistance of line management (James E. Stewart and René Lavoie of DFO and Roger Foxall and Don Robson of the NRC). Finally, we must thank our wives, Ingrid and Patricia, and our families for their patience, support and understanding during the many late nights and weekends devoted to the project. Although we have taken great pains to eliminate errors, any project of this size will contain errors of both omission and commission. We would be grateful to have such lapses called to our attention.

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August 1987

INTRODUCTION

Interest in ozone has increased dramatically in recent years. Ozone has moved from relative obscurity to celebrity status, chiefly because of its interrelationship with pollution and the ecological consequences to man. As this introduction is being written, the international news media are headlining stories on stratospheric ozone depletion over the antarctic region of our globe, the depletion possibly related to man's chemical pollution. The worldwide interest in ozone is being expressed as a cornucopia of published scientific studies.

Interest in aquatic applications of ozone has also increased drastically. Initially applied mainly for drinking water sterilization, ozonation is now widely used in municipal waste-water treatment, in process-water polishing, in soft-drink manufacturing, in pulp and paper waste-water treatment and many other industrial applications. During the past decade, aquaculturists have become interested in using ozone for a number of purposes, including sterilization of intake and effluent water, quarantine waste-water treatment, and water treatment in recirculation systems where ozone is used to reduce the organic load and to control bacterial counts.

One exciting new prospect in the fresh-fish industry is the use of ice containing residual ozone. As the ice melts, it releases ozone which destroys decomposition bacteria on the fish, thus extending the shelf life of the fresh fish by up to 48 hours. Though promising, this technique is still only in the developmental stages. However, similar techniques have been used in other branches of the food-processing industry. For example, Shimizu *et al.* (1982) report that in grape processing, a 30-minute immersion into ozonized water followed by air-tight packaging is effective in delaying grape rot in cold storage. Attempts have also been made to apply ozone as a means of detoxification for red tide toxins in the shellfish industry. Gukalina *et al.* (1983) report on extended storage of potatoes through ozone treatment and Kaess (1956) describes the effects of ozone on chilled meat. Yakushi (1983) reviews advantages and disadvantages of food sterilization and processing by ozone.

With its growing number of applications in fresh, brackish and marine waters, research on ozone has addressed various basic questions in recent years. These include ozone generation, its optimization and determination in various media, general ozone chemistry and ozone toxicity. In the past, most of the information available in these areas originated from other fields of interest. The bulk of information on toxicity and biological effects stemmed from environmental studies with plants, while most of the data concerning effects on materials were generated with the plastic industry. More recently, in conjunction with rising concern about the effects of atmospheric pollution and ozone/oxidant interactions, a wealth of information on ozone toxicity has been collected through physiological and biochemical investigations using vertebrates. Medical studies have concentrated on lung and blood physiology at ozone-exposed workplaces.

The recent literature on ozone is, therefore, widespread and not easily accessible to scientists and practitioners working in the aquatic disciplines. In an attempt to facilitate better access to this literature, a *"Selected Bibliography on Ozone, its Biological Effects and Technical Applications"* was prepared during the early 1970's (Rosenthal, 1974). At the time, this bibliography contained only a few references dealing with aquaculture. Shortly after its appearance the rising interest in ozone required an early reprint of the bibliography, although it was soon outdated. During the last few years an increasing number of publications have appeared which describe the utility of ozone in aquaculture. Additionally, the literature dealing with ozone chemistry in aquatic media and with its effects on aquatic organisms is rapidly expanding. It was therefore felt useful to compile an updated version that allows scientists and practitioners to orient themselves quickly in this relatively new field of ozone application while at the same time providing the opportunity to access the information accumulated in related fields.

The current bibliography attempts to marshall the available ozone literature in the following fields:

1. Generation of ozone. (Subject Index, Section 2).
2. Determination of ozone in both air and aquatic media. (Subject Index, Section 3).
3. Application of ozone to water and waste-water treatment in both municipal and industrial facilities. (Subject Index, Section 6).
4. Utilization of ozone in home aquaria, public aquaria, fish hatcheries, shellfish purification units, recirculation systems and other aquaculture installations such as quarantine stations. (Subject Index, Section 6.15).
5. Properties of ozone and its chemical reactions, especially in aquatic media. (Subject Index, Sections 4 & 5).
6. Biological effects of ozone on viruses, bacteria, invertebrates, vertebrates and man as well as algae and plants. (Subject Index, Section 7).
7. Effects of ozone on materials. (Subject Index, Section 8).

Additionally, the most important publications on ozonation products, degradation products of ozone-oxidized organics and reaction products with inorganics as well as the biological effects of such products have been included (Subject Index, Sections 5 & 7). Further, the growing literature on antioxidants and antiozonants to protect both materials and living organisms against the oxidizing capacity of ozone has been selectively treated (Subject Index, Section 7.3). Finally, we include a portion of the voluminous information available on air pollution and natural atmospheric ozone phenomena (Subject Index, Section 1).

ORGANIZATION OF THE BIBLIOGRAPHY

The bibliography consists of 4775 references in alphabetical order by author name, with 16 additional citations at the end of this introduction, 4791 *in toto*. The references date from 1845 to mid-1986, with a few entries from late 1986 or early 1987. About 80% of the references are from 1965 or later, and about 55% of the references are later than 1975. This is illustrated in Fig. 1, a histogram showing the number of references per year in this bibliography. The apparent drop-off in ozone publications in 1985 and 1986 is artificial, caused by our cut-off date for entry of new material in mid-1986.

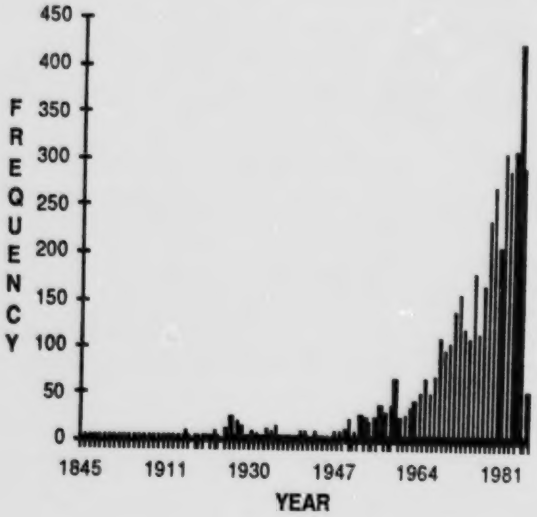

FIG. 1. Frequency occurrence by year of references included in this bibliography.

The bibliography consists of two sections: first, the Bibliography proper, a listing of the citations alphabetically by author name, and secondly, the Subject Index, a reference index with all the citations sorted by subject.

Within the Bibliography proper, each citation follows the format: Serial Number- Author- Date- Title- Publication Name- Volume Number- (Issue Number)- Pages, with each entry assigned a sequential serial number. At the end of each reference, the original language of publication is indicated if other than English. The title of the citation has usually been translated into English. Often these title translations differ from one citation source to another, so in many cases we have also provided the title in the original language. Immediately below each citation, we have also provided, if possible, one or more cross-references to the various abstracting services where an abstract of the cited publication may be found. This permits interested researchers to easily locate an abstract of the article if they want to evaluate it before trying to obtain the original publication. Where possible we have tried to include abstract citations from the two most widely available abstracting services, Biological Abstracts and Chemical Abstracts. Citations from other reference services such as Aquatic Sciences and Fisheries Abstracts, Water Pollution Abstracts, Dissertation Abstracts, *etc.* are frequently included.

The Subject Index consists of a Category List and the Main Index. The Category List is provided to briefly show the organization of the subjects in the Main Index. In the Main Index, each of the subject category headings is followed by a list of reference entries in which each of the entries consists of the citation serial number from the Bibliography as well as the author name and date of publication. This is somewhat redundant but we feel from personal experience that reference lists consisting of long columns of numbers are anathema, and we much prefer to see a listing of names. It is our experience that workers in a particular field will probably know many of the citations by author name and date and will need to look up only those with which they are not familiar. In the interests of conciseness, the citations are indexed in one or two categories only. Extensive cross-referencing was avoided to restrict the number of entries per category. We feel this encourages the use of the Subject Index in that users are not confronted with hundreds of references for each subject.

This edition of the bibliography, unlike the 1974 edition, was done on microcomputers. The project began in Germany on a custom computer assembled from component parts, using dBase II (Ashton-Tate) for data entry. In Canada the project continued using an Apple IIc computer equipped with a Z-80 CP/M card (Applied Engineering), also using dBase II for data entry. The database was then transferred from the Apple IIc using Wordstar (MicroPro) for data output to an Apple Macintosh computer with a 20 Mb internal hard disk (Hyperdrive) using Versaterm (Peripherals) for the data capture. On the Macintosh the data were formatted using

PortaAPL (Portable Software) and finally edited using Word (Microsoft). Final printing was done on an Apple Laserwriter. The complete bibliography occupies about 3 double-sided 3-1/4 inch diskettes of 800 K each.

TOPICS OF SPECIAL INTEREST

It is not the intent of the authors to provide a comprehensive review of the progress made in any of these fields since the 1974 edition of this bibliography. However, a few aspects will be highlighted which are considered to be of special interest to ozone researchers and practitioners, particularly aquaculturists and aquatic biologists. These brief notes can mention but a few of the many recent important contributions to the scientific knowledge of ozone, the pale blue gas with so many important properties.

OZONE PREPARATION AND GENERATION

Since 1974 a landslide of publications on ozonizers and ozone generation has appeared, with numerous procedures awarded patents, predominantly in Japan, but also in the USSR, the US, the UK, France, Germany and other European countries. Many, but not all, of these recent patents have been listed in this bibliography.

The performance of ozonizers has been drastically improved in recent years and test results on the effectiveness of ozone output under a variety of operational conditions are now available. For example, the wave form of the applied voltage was found to affect electrical properties of modern discharge ozonizers, but not the kinetics or energy yields in ozone formation (Vorozhtsov *et al.*, 1983). Many recent studies have determined the extent of nitrogen-oxide formation in ozonizers using ambient air as a gas source (*e.g.*, Samoilovich *et al.*, 1984). With increasing yield of ozone in discharges, the oxidation state of nitrogen increases. Firstly, NO is formed, then NO_2 or N_2O_5 and to some extent NO_3. A number of studies have demonstrated how to improve energy efficiency in ozone generators, while operating costs as well as maintenance costs have been greatly reduced. The development of special electrodes has helped to improve energy efficiency of ozone generators by more effective use of amplitude modulators, with uniform charging of multiphase circuits, employing pulse-phase control units. Output efficiencies between 17 and 25 g O_3 per Wh are frequently reported.

OZONE DETERMINATION AND MEASUREMENT IN AQUATIC MEDIA

At present about 10 analytical methods are available for ozone determination in aquatic media. Although most of the publications mention residual ozone as their target determinant, they mainly provide techniques to quantify total radical oxidant present in aqueous solutions. Four of the methods are iodometric techniques that are based on the reduction of ozone by iodide ions. They include the standard iodometric method, the amperometric method, the arsenic (III) back titration method, and the DPD method. The remaining methods include direct UV absorption at 259 nm (Bahnemann and Hart, 1982; Forni *et al.*, 1982), the acid chrome violet (ACVK) method (Masschelein and Franselot, 1977), a membrane electrode (Stanley and Johnson, 1979), the indigo method (Bader and Hoigne, 1981, 1982), the bis(terpyridine) iron (II) method (Tomiyasu and Gordon, 1984) and a gas diffusion flow injection analysis (Straka *et al.*, 1985). Detection limits have been improved in recent years. Several methodological modifications have reduced significantly the chlorine interference, which is important to those who use ozone in brackish and seawater. Intercalibration practices between several methods have demonstrated the improved reliability of these techniques and have helped to remove a number of earlier methodological inconsistencies.

REACTIONS OF OZONE WITH INTERFERING GASES

Much progress has been made in studying the kinetics and reactions of ozone with other gases such as nitrogen dioxide and sulfur dioxide. Most of the literature in this area originates from air pollution studies. For example, Evans *et al.* (1984) describe methods of correcting spectrophotometer measurements of total atmospheric ozone in order to compensate for interference caused by atmospheric sulfur dioxide. Schenkel and Broder (1982) study the effect of trace gas interference (NO_2, SO_2, NO & CO) on ozone measurement via the potassium iodide method and they give theoretical interpretations for the effects of the gases. Much can be learned from these and similar investigations when considering the reactions of gases with ozone in aquatic media. It is for this reason that selected references on ozone and air pollution are included in this bibliography.

DECOMPOSITION OF OZONE IN AQUEOUS SOLUTIONS

Decomposition of ozone in water has been extensively studied in the past. Most of the investigations are largely based on phenomenological descriptions of overall kinetics found for "pure water". Most systems are kinetically complex because a primary decomposition of ozone produces free radicals which may either be scavenged by bicarbonate and organic solutes or which may react with further ozone to yield more free radicals, thus accelerating the decomposition of ozone (Staehelin and Hoigne, 1982). Although various studies have been undertaken in recent years, the kinetics of such chain reactions are not yet well understood, mainly because of the many parameters involved. Major aspects of this problem have been investigated and reviewed by Stumm (1954), Peleg (1976), Roth *et al.* (1979), Gurol and Singer (1982, 1983) and Staehelin and Hoigne (1983). Recently, Staehelin and Hoigne (1985) demonstrated the chain reaction in ozone decomposition involving OH^- radicals. Many organic solutes (impurities) can react with OH^-, some acting as promotors, others as OH^- radical scavengers that do not primarily produce O_2. Further studies are expected to provide quantitative interpretations for the observed variation of the lifetime of ozone found in many test solutions and natural waters.

CHEMICAL REACTIONS OF OZONE AND AQUATIC APPLICATIONS

The strong electrophilic nature of ozone imparts to it the ability to react with a wide variety of organic and organometallic functional groups. The vast majority of ozone reactions are based on the cleavage of the carbon-carbon double bond. The inorganic chemistry of ozone involves nearly all members of the periodic chart. Ozone does not react with metal ions which exhibit only one oxidation state, *e.g.*, calcium and sodium, but it may react with other metals to form ozonides and oxides or both.

All members of the halide series, except fluorine, react with ozone. Fluorine does not react with ozone because it is the only element that has a higher oxidation potential than ozone. Bromide is quantitatively oxidized with ozone to bromine, and iodide reacts with ozone to form iodine.

In brackish and marine waters the chemistry of ozone is dominated by its reaction with Br^- due to bromide's relatively high concentration (65 mg/L) and reactivity compared with other seawater compounds. Ozone oxidizes Br^- under water treatment conditions to form HOBr. The HOBr reacts further with ozone, but only in its ionized form, OBr^-. The OBr^- is oxidized not only to BrO_3- but also to a species that regenerates Br- (Haag and Hoigne, 1983). In the presence of organic matter, HOBr reacts to form bromo-organics. However the range of conditions conducive to haloform formation is narrower than during chlorination. (Stewart *et al.*, 1979).

Ammonia oxidation in aqueous solutions by ozone is slow compared with that of nitrite. Ammonia oxidation in water can be initiated by ozone in two ways (Hoigne and Bader, 1978): (a) direct reaction of ozone with NH_3 or (b) reactions of OH^- radicals formed upon O_3 decomposition. The direct reaction seems to predominate at pH levels above 9. Whenever the OH^- mechanism is involved, NH_3 is easily protected by other solutes which also consume OH^- radicals. Even carbonate and bicarbonate ions may act as efficient OH^- scavengers, which explains in part the very slow oxidation of ammonia in seawater. Singer and Zilli (1975) concluded from their studies with ammonia-containing municipal waste waters that ozonation alleviates the oxygen-demanding characteristics, transforming ammonia to nitrate in a first-order reaction, catalyzed by OH^- over the pH range 7-9. However, ammonia competes for ozone with the dissolved organic constituents comprising the COD, but is oxidized preferentially provided alkaline pH values can be maintained. Due to the elevated pH's required, ammonia oxidation by ozone seems attractive only in conjunction with lime clarification and precipitation of phosphate.

Nitrite oxidation can be very effectively achieved by ozonation. This holds not only for waste-water treatment but also for process engineering. For example, $NaNO_3$-containing electrolytes used for electrochemical machining of iron-containing materials are partially reduced, thereby releasing large quantities of nitrite. Ozone is applied as an oxidizing agent, permitting the complete recycling of the electrolyte. This is due also to the simultaneous oxidation of iron to iron hydroxide which precipitates easily and renews the electrolyte to its original composition (Lindner *et al.*, 1984).

The treatment with ozone of activated carbon used as filter media modifies the distribution of the porous volume of the carbon, creating surface conditions which appear to affect the rate, capacity and character of pollutant elimination. The oxidation favors the retention of adsorbable and non-adsorbable molecules of differing sizes and permits a more effective biological treatment (Gaid *et al.*, 1983; Guirguis *et al.*, 1978).

Gilbert (1984) has shown that the low biological degradability of various toxic and/or refractory substances can be converted, by ozonolysis, to biologically active groups of substances in some cases but not in others. The reaction of aniline, for example, with low ozone doses forms products that are not biodegradable. In many cases the intermediate ozonation products are scarcely easier to degrade than the initial substance.

Ozonation of dyes substantially increases their biodegradability for waste-water treatment. In a recent study Matsui *et al.* (1984) could demonstrate that the total organic carbon (TOC) content and the BOD of the ozonation products agreed well with the theoretical TOC values of the original dye. Azo-dyes were easily decomposed.

One interesting observation relates to the effect of ozone on the elimination of various algae during filtration (Ginocchio, 1981). Algae were effectively removed from lake waters by ozonation and flocculation-filtration in dual filter media.

CONTACTING DEVICES

The number of studies concerning the design of contacting systems to apply ozone in aquatic media has increased drastically over the last few years, as a result of recognition that intensive mixing between ozone (or ozone-containing air) and the solution to be treated play an important role in achieving efficient utilization of this powerful oxidant. Most of the past studies in this field provided inadequate data for process optimization because of insufficient differentiation between the parameters for mass-transfer, contact time and reaction time. In the future, detailed evaluations of gas-liquid contacting devices are necessary so that ozone utilization can be optimized and operating costs minimized. Sheffer and Esterson (1982) have provided a detailed study that examined the mass transfer and the reaction kinetics of ozone in tap water, and derived a mathematical model describing the mass transfer characteristics in a reactor for various operational parameters. These studies confirmed and improved earlier findings (Hill and Spencer, 1975; Hewes and Davison, 1971). Gurol (1985) provided a model for estimating the effectiveness of organic removal in ozone reactors in relation to various operational conditions of these reactors.

OZONE APPLICATION TO CULTIVATION OF AQUATIC ORGANISMS

Aquaria Systems

Murphy (1975)* favoured ozone application for closed marine aquaria systems because of (a) removal of colour which normally taints aquarium water, (b) reduction of BOD and COD in culture media and (c) reduction in bacteria levels. Sander and Rosenthal (1975) showed that redox potential of sea water in large aquarium systems was substantially enhanced by ozonation. Rosenthal and Fonds (1973) and Fonds (1970) applied ozone as an additional means of water treatment in laboratory rearing trials using larvae and fry of marine fish. Moffett and Shleser (1975)* tried successfully to rear first-stage lobster larvae (*Homarus americanus*) in ozone-treated water at a final dissolved ozone dose of 0.4 mg/L for a treatment period of 30 minutes. Then, the sea water was recirculated for six days without additional ozonation and lobster larvae survived at a rate of about 75%.

Honn and Chavin (1976) studied the utility of ozone treatment in the maintenance of water quality for closed systems of public aquaria. Honn (1979) describes the maintenance of filter-bed effluent at low levels for total ammonia, un-ionized ammonia and nitrite when ozone was used as an oxidative supplement to biological filtration in both small-scale (2.27 m^3) and large-scale (75.7 m^3) systems. Within 24 hours of the cessation of ozonation these levels were reversed (Table 1). During normal operation of the closed system, bacterial counts were similar in the aquarium and the algal-bed bacterial-filter effluents, but were significantly reduced by ozonation. Termination of ozone treatment resulted in a significant increase in the reservoir effluent bacterial number after 3 hours.

Honn (1979) concluded from his 18-month evaluation of the marine recycling system that while nitrogenous waste content levels of the sea water were highly variable, that nevertheless, levels were maintained within acceptable limits and this was attributed to the flexibility imparted to the system with ozonation. The variable levels were not surprising considering that no attempt was made to maintain a constant biomass. Reservoir ozonation consistently reduced total ammonia levels to within acceptable limits.

TABLE 1. Effects of termination and resumption of ozonation on nitrogen sources in the aquarium and reservoir effluent of a marine aquarium recycling system (modified after Honn, 1979).

OZONATION TERMINATED (Hours)	T_0	T_6	T_{24}
Aquarium effluent			
Total NH_4	0.24	0.30	0.59
Total NO_2	0.47	0.51	0.83
Reservoir effluent			
Total NH_4	0.04	0.16	0.30
Total NO_2	?	?	?

OZONATION RESUMED (Hours)	T_0	T_6	T_{24}
Aquarium effluent			
Total NH_4	0.31	0.20	0.27
Total NO_2	0.78	0.48	0.34
Reservoir effluent			
Total NH_4	0.07	0.03	0.03
Total NO_2	0.22	0.11	0.04

Ozonation can support stability in water quality, especially in recycling systems (see Rosenthal, 1981). The importance of maintaining a high water quality in intensified fish cultivation is stressed by Wedemeyer (1981)* when comparing fish health of rainbow trout reared in recirculating and flow-through systems at the same hatchery. Although the recirculation system produced larger smolts, these fish suffered from several physiological abnormalities.

There are several observations reported in the literature that indicate ammonia oxidation in aqueous solution, although the reaction is fairly slow (Hoigne and Bader, 1978; Singer and Zilli, 1975; Huibers *et al.*, 1969; Rosenthal *et al.*, 1978).

A combination of ozone treatment with countercurrent foam stripping seems to hold some promise for aquacultural applications. Schlesner and Rheinheimer (1974) investigated the effectiveness of ozonation units with regard to bacterial counts in treated recirculating water of the public aquarium in Kiel, Germany. They found a considerable reduction in saprophyte plate counts in both the brackish water and full strength seawater system. Since the waste water was considerably loaded with organic wastes, they attributed the decrease of the bacterial count to the fact that ozonation was combined with countercurrent foam stripping. Ozone had a rather indirect effect due to its oxidizing capacity. An increase in surface tension caused by smaller air bubbles in the reaction chamber resulted in a more consistent foam production. This foam acted as a collecting agent for bacteria, aggregating them considerably in the foam. Since the ozonation unit was operated at only half of its initial capacity and with ozone added to only one of its reaction towers, the ozone concentration maintained in the treatment unit was too low to allow complete disinfection. Other reports on improvements in combined ozone treatment with countercurrent foam stripping devices for fish farm waste-water processing are presented by Rosenthal and Kruner (1985).

Pilot-Scale Recycling Systems

There are several trials using ozone in fish culture recycling systems. Morrison *et al.* (1979) investigated the effects of a pilot-plant ozonation unit at the Dworshak National Fish Hatchery on the efficacy of sterilizing make-up water entering the experimental recycling systems of the hatchery. Analysis of plate counts showed that ozone consistently provided better disinfection of the make-up water than the existing ultra-violet system. Additionally, ozone destroyed unicellular algae which ultraviolet light did not. Ammonia levels showed a 70% decrease with ozone sterilization. When including the ozonation unit in the system, BOD load showed no significant increase or decrease, but there was a considerable levelling effect during the ozone run. The authors considered it as most important that biofilter performance could be enhanced by ozonation, causing a general decrease of stress on the fish.

In recent years, detailed studies on the effectiveness of ozone treatment in intensive fish culture recycling systems were undertaken by Otte *et al.* (1977), Rosenthal *et al.* (1978, 1979) and Rosenthal and Otte (1979). When applying ozone in a bypass system with combined foam stripping, ammonia concentrations were slightly reduced in about 72% of the treatments.

Singer and Zilli (1975) demonstrated in municipal waste water a dependency of ammonia oxidation with ozone on pH levels. Effective first-order reaction with respect to ammonia concentration can be maintained only in alkaline medium. These authors observed a rate increase with increasing pH over the range 7 to 9. Due to the elevated pH required for effective ammonia oxidation, ozonation seems especially attractive to be included in brackish and sea-water treatment procedures.

In the experimental recycling system, nitrite oxidation by ozone was dependent on the ozone concentration employed. Ozone concentrations of 2-4 mg/L resulted in an average reduction of about 50% of the initial nitrite concentration. Higher ozone dosages led to an almost quantitative oxidation of nitrite to nitrate.

Ozone treatment of recirculated water had a remarkable effect on the content of low biodegradable organics in the culture medium. At ozone concentrations between 6.8 and 10.4 mg/L the BOD_5 load of the treated water increased substantially, indicating that those substances not easily attacked by the bacteria of the biofilter are cracked into smaller pieces which are then available for biological degradation. The effect of ozonation on low-degradable organics is more clearly demonstrated when determining COD and TOC degradation in the system at times with and without ozonation (Rosenthal and Kruner, 1985). Without ozone treatment, substances highly resistant to biological decomposition accumulate in a recycling system. Intermittant ozonation proved to yield the best results in cracking those substances not easily digested by biofilters.

Similar observations are reported by Mallevialle *et al.* (1978), who studied the destruction of humic substances by ozone under various experimental conditions, such as contact time, color, total acidity and the concentration of polyhydroxyaromatic derivatives. These authors found color removal of more than 90% within 10 minutes.

Continuous decrease in TOC showed that there was a gradual oxidation of the organic compounds. Further, during the initial depolymerisation phase, the BOD5 increased, which confirms the observations by Rosenthal *et al.* (1979) on the formation of more biodegradable substances. There exists also other experimental evidence that ozonation cracks long-chain organics dissolved in waste water. For example, the quantity of organic acids in Lake Constance water increased from 60% to about 72% of the total BOD load when ozonation was applied, whereby substantial amounts of soluble organics were transferred to more polar substances with smaller molecular weights (Maier, 1974), a fact which can be utilized when combining ozone treatment with foam fractionation.

TABLE 2. Examples of marine aquarium systems using ozone as a means of water treatment.

Total water volume	Ozone output	Other treatment facilities	Types of animals
Sea World (Ohio, USA)			
450,000 gal	20-40 lb/day	High-rate sand filter	Marine mammals
Sea World (Florida, USA)			
2,000,000 gal	70-100 lb/day	Gravel filter, gravity feed	Marine fish sharks, turtles
167,000 gal	1.0-1.5 lb/day	" " " "	" " " "
Kiel Aquarium (FRG)			
129,000 L (Brackish)	17 g/hr	Aerated algal tank, mech.	Baltic species
148,000 L	17 g/hr	" " " "	North Sea fishes
Biologische Anstalt Helgoland (Hamburg, FRG)			
8,500 L (Brackish)	15 g/hr	Trickling filter, settling tank, denitrification	Warm-water fishes
Epcot Living Seas (Florida, USA)			
6,000,000 gal	35 lb/day	High-rate sand	Reef fish, mammals, sharks, turtles

In several public aquarium systems ozonation is successfully applied for water treatment (Table 2). For example, in the United States, the Sea World oceanaria in Ohio and Florida ozonize large quantities of recirculating sea water continuously (Murphy, 1975).* Walt Disney's Epcot Center, Orlando, Florida, has built the world's largest marine aquarium. This facility uses ozone for colour removal, disinfection and organic oxidation. This unique aquarium (6 million U.S. gallons) mixes fish, invertebrates and marine mammals in a common water system. Sea World of Ohio exhibits only marine mammals. Their waste water passing from the exhibition tank is ozonized in a contacting system and passes through a sand filter after which it is returned to the tank. At the Sea World in Florida comparable systems are in use. Blogoslawski *et al.* (1975) reported the design of a sea-water filtration system which uses ozone. It consisted of a static contacting device for optimal mass ozone transfer and activated carbon filters which removed oxidized organic material as well as residuals resulting from the ozonation of seawater.

Shellfish Purification and Quarantine Stations

Chlorination has been employed for many years in oyster purification plants. This process is often called shellfish depuration or cleansing. During the early 60's, however, the trend was toward the development of cleansing plants which use other means of water purification. Since chlorination is not considered to play an important role in water sterilization for aquaculture purposes, this method will not be considered in detail. The earliest trials to utilize ozone in sterilizing sea water to which human pathogens had been added was demonstrated by Violle (1929). In France (Anonymous, 1972) and later in other areas, practical application of ozone led to the development of commercial ozone depuration stations for shellfish. For many years, this process had successfully been employed for cleansing shellfish contaminated with coliform bacteria (Salmon *et al.*, 1937*a*, *b*). The experimental results offered by Fauvel (1963) served as a guide for the construction of the existing 9 depuration installations in France (Blogoslawski, 1977). The older stations required one month to cleanse the contaminated animals, whereas those stations employing ozone can depurate contaminated shellfish within 48 hours.

Ozone disinfection of the water supply in a mariculture facility ensures not only the prevention of diseases caused by contaminated water, but also serves as a quarantine measure to protect the environment surrounding a hatchery from transfer of pathogens introduced with exotic species. For example, the Prince Edward Island Department of Fisheries (Canada) introduced bay scallops (*Argopecten irradians irradians*) into their province for commercial cultivation in 1981. Considering the fact that bay scallops are not indigenous to the area, a quarantine period was deemed essential for the protection of the native marine environment. This period continued with regular pathological testing until a disease-free state was ensured. Shellfish depuration and public health was reviewed by Blogoslawski and Stewart (1981).

Disease Treatment

Colberg and Lingg (1978) suggested using ozone not only as a disinfectant for make-up water in fish hatcheries but also for destroying bacterial fish pathogens in recycling systems, since increased levels of metabolites do accumulate in such systems and enhance the potential for outbreak of diseases. These authors established mortality rates for four bacterial fish pathogens (*Aeromonas salmonicida*, *A. liquefaciens*, *Pseudomonas* *fluorescens, Yersinia ruckeri*) during batch and continuous flow ozonation. Over 99% mortality of the fish pathogens was observed within 60 seconds of contact during continuous flow exposures at 1.0 and 0.1 mg O_3/L. According to Colberg and Lingg (1978), *Aeromonas salmonicida* was the most sensitive to ozone in the continuous flow ozone studies, which is contrary to data of Wedemeyer and Nelson (1977) who reported that *A. salmonicida* was more resistant to ozone than was the causitive agent of enteric redmouth disease (ERM). Although these differences are largely unexplained, one may assume that factors such as strain differences, pH differences, and differences in culture conditions (temperature, pre-treatment of media) affect the response to ozone exposure. In phosphate-buffered distilled water, 0.01 mg /L ozone inactivated 103 cells/mL of ERM and *A. salmonicida* in 0.5 and 10 minutes, respectively. Ozonation of untreated lake water at 90 mg O_3/L/h (equivalent to 0.01 mg/L residual in ozone-demand-free water) was required to destroy both pathogens within 10 minutes.

Wedemeyer *et al.* (1978) determined ozone inactivation curves in three water types at 10 degrees C for the fish pathogenic viruses, infectious hematopoietic necrosis (IHNV) and infectious pancreatic necrosis (IPNV). In phosphate-buffered distilled water an ozone dose of 0.01 mg/L for 30 or 60 seconds inactivated IHNV or IPNV, respectively. In hard (120 mg/L as $CaCO_3$) and soft (30 mg/L) lake waters, an ozone application rate of 70 mg/L/h for 10 minutes destroyed IHVN. Inactivation of IPNV in hard water required 90 mg O_3/L/h for 10 minutes, but only a 30-second contact-time in soft water. Based on these results and comparable investigations with chlorine, Wedemeyer *et al.* (1978) recommended the use of ozone as a disease control agent in fish hatcheries. Conrad *et al.* (1975) and Rosenlund (1975) also studied ozone as a nonpersistent disease control agent in intensive fish culture in order to destroy bacterial and viral fish pathogens in hatchery influent and effluent waters. Conrad *et al.* (1975) showed that ozone effectively destroyed *Flexibacter columnaris* at 103 to 105 cells/mL but unfortunately the effective contact time and actual dose had not been determined.

Burleson *et al.* (1975) studying the effects of ozone on human pathogens, included one fish pathogen in their investigations: *Pseudomonas fluorescens*. They found complete and rapid destruction (within 1 to 2 minutes) at relatively low, but variable, ozone concentration when *Pseudomonas fluorescens* was suspended in secondary sewage treatment effluent at about 10^7 cells/mL.

Straub (1975) reported on successful destruction of bacteria in hatchery supply water by ozone, thereby drastically enhancing survival of eggs. Mortality reached up to 45% of the total number of eggs incubated in untreated water, whereas egg losses in ozonized water were only 4%-5%.

Danielsen (1975)* studied the effect of seawater ozone treatment in a flow-through system for coho salmon smolt production. As a direct result of the ozone treatment of sea water, disease mortalities were expected to be substantially less in the test group than in the control group. Ozone was applied at a rate of 3 g/h and water flow rates were maintained at 5 gpm. Residual ozone was removed by an activated charcoal filter before the water entered the rearing tank. Because no epidemic of vibriosis was encountered during the experiment, the initial hypothesis was not proven. However, there was evidence that better growth had been achieved by the test group.

Detoxification of Red Tide Toxins and Other Marine Poisons

Red tides, or blooms of toxic dinoflagellates, occur in marine waters throughout the world. In many cases these organisms accumulate in molluscs, not being harmful to these animals themselves, but toxic to those vertebrates eating them. The ingestion of poisonous shellfish has been responsible for many human and other mortalities. Dawson *et al.* (1976) found that extracts from *Gonyaulax tamarensis*, containing the toxin which causes paralytic shellfish poisoning (PSP), could be detoxified by ozone treatment. Mussels held in raw seawater from the red tide area had built up high levels of toxicity, while those held in ozonized red tide seawater remained non-toxic.

Ozone proved also to be capable of inactivating botulism toxin (*Clostridium botulinum*) (Miller, 1959, Graikoski *et al.*, 1984) and *Gymnodinium breve* toxin, a toxic red tide metabolite (Blogoslawski *et al.*, 1973). Thurberg (1975) tested the inactivation of of red-tide toxins by ozone treatment, using isolated toxins of three dinoflagellates (*Gymnodinium breve, Gonyaulax catanella,* and *Gonyaulax tamarensis)*. After ozonation, solution samples were injected into mice and killifish in order to evaluate the ozone-induced loss of toxicity. The impressive results are summarized in Table 3.

The metabolites produced by some red tide organisms present potential water quality problems to those facilities which depend on seawater supplies for maintaining or cultivating marine organisms. Thurberg (1975) reported on shellfish beds closed for harvesting by local officials in September 1974, when paralytic shellfish poisoning was building up in clams and mussels after a *Gonyaulax tamarensis* bloom. The results of recent investigations indicate some hope for the applicability of ozone in pre-treatment of seawater for those coastal shellfish farms or purification units which are frequently exposed to red tide waters. A number of these studies show that inactivation of the toxins can be achieved in some cases (Blogoslawski and Stewart, 1978; Blogoslawski *et al.*, 1973, 1975, 1979). However, further research is needed before introducing ozone treatment procedures on a large scale. Recent investigations (White *et al.*, 1985) with soft shell clams (*Mya arenaria*) show that ozone is ineffective when high concentrations of ozone were employed because the clams pumped poorly under ozone exposure. Results indicate the ineffectiveness of ozonation in detoxifying soft-shell clams at least when they have retained the toxins for long periods.

TABLE 3. Ozone inactivation of dinoflagellate toxin as determined by mouse bioassay using a 5-minute exposure to 2 % ozone gas at different flow rates (modified after Thurberg, 1975). G. b. = *Gymnodinium breve*, G. c. = *Gonyaulax catanella*, G. t. = *Gonyaulax tamarensis*.

Ozone flow mL/min	No. of mice	Toxin source	48-hour survival %
110	10	G. b.	100
65	10	G. b.	80
40	5	G. b.	40
20	10	G. b.	0
110	5	G. t.	100
55	10	G. t.	90
27	10	G. t.	20
0	20	G. t.	0
110	15	G. c.	100
55	15	G. c.	100
27	10	G. c.	0
0	20	G. c.	0

Ozone detoxification of other biologically active marine compounds such as Tetrodon poison has been tried by Blogoslawski and Stewart (1977). Tetrodon poison, also known as fugu or puffer fish poison, is caused by tetrodotoxin (TTX).

Many other investigations by various authors confirmed a greater effectiveness of ozone in water sterilization when comparing the differences between chlorination and ozonation (Hann, 1956; Kessel *et al.*, 1943, 1944; Lagrange and Rayet, 1952; Bringmann, 1954; Fetner and Ingols, 1956; Lue-Hing *et al.*, 1977; Namie, 1977).

BIOLOGICAL EFFECTS AND HEALTH HAZARDS

There is a tremendous amount of literature available on the biological effects of individual air oxidants such as ozone and the combined effects of individual pollutants with ozone on members of the plant community such as beans, wheat and pines. Most of the studies look at impaired growth, leaf injury, reduced disease resistance and specific aspects of biochemistry mainly in commercially important plants such as beans (*e.g.*, Hofstra and Beckerson, 1981; Hucl and Beversdorf, 1982; Ito *et al.*, 1985), potatoes (Hofstra *et al.*, 1983; Illman *et al.*, 1981), rice (Kats *et al.*, 1985), and other species. Since our interests are principally aquatic, we do not intend to review this material here although many of these references are included in this bibliography.

A number of recent studies include the evaluation of physiological responses of various species to ozone exposure, such as altered structures of immunoglobulin cells (Gershwin *et al.*, 1981), reduced collateral resistance (Gertner *et al.*, 1983), inhibition of tissue cholinesterase (Gordon *et al.*, 1981), the pharmacokinetics of pentobarbital (Graham *et al.*, 1985), the lecithin metabolism (Ichikawa *et al.*, 1982), the effects on hepatic microsomal enzymes (Graham *et al.*, 1982), the degradation of DNA (Hamelin *et al.*, 1978), the fatty acid metabolism (Grunwald and Endress, 1984) and the isoleucine metabolism (Harada *et al.*, 1983).

Viruses and Bacteria

The use of ozone for inactivation of viruses has increased. Pioneering work leading to the formulation of a relationship between the number of organisms present at any time after exposure to the disinfectant has indicated that, for example, enterovirus inactivation by ozone does not follow single-order kinetics (Roy *et al.*, 1981, 1982). The cause of such deviation appears to be the inherent properties of the virus (medianistic concept) rather than the heterogeneity of the virus population (vitalistic concept). Roy *et al.* (1982) provide an empirical model which resembles the exact survival equations to allow prediction of ozone inactivation of poliovirus within +/- 0.5% accuracy.

Harakeh (1984) investigated the combined effects of various disinfectants against poliovirus 1 in a municipal waste-water effluent and found that the efficacy of chlorine was enhanced in the presence of ozone.

Further studies by Harakeh and Butler (1984) provided evidence that the inactivation of the enteric viruses in effluents was most effective at low pH and the presence of suspended solids had no substantial effect on inactivation, but pre- or post-treatment by ultrasound enhanced inactivation.

Rapid inactivation of the bovine renal tracheitis virus was demonstrated by Ancieux *et al.* (1982), provided the virus was still suspended in saline solutions. The virus was not inactivated by ozone after absorption to cells.

Bolton *et al.* (1982) exposed representative viruses from five major virus groups to moderate ozone concentrations in order to establish the biological relevance of the reaction of ozone with soluble proteins and lipid biolayer membrane systems. The order of susceptibility to ozone inactivation of the enveloped viruses was vesicular stomatitis virus (VSV) (Rhabdoviridae), influenza A virus (WSN strain) (Orthomyxoviridae), and infectious bovine rhinotracheitis virus (IBRV) (Herpesviridae). In contrast to the enveloped viruses, the non-enveloped viruses were relatively resistant to ozone inactivation.

Newton and Jones (1949) observed a 99% kill of *Endamoeba histolytica* cysts within 1 to 3 minutes in aqueous solutions containing 0.5 to 1.0 mg/L ozone. This cysticidal action did not appear to be influenced by pH, temperature, or organic nitrogen content of the water. As pointed out by Venosa (1972), *E. histolytica* cysts are relatively resistant to chlorine.

Extended studies on the effectiveness of ozone treatment of bacteria in both tap water and raw stream water were performed by Dickerman *et al.* (1954). The organisms tested were staphylococci, enterobacteria, bacilli, and pseudomonads. In every instance, except for *Bacillus subtilis*, application of ozone to produce a residual concentration of 2 mg/L was sufficient to kill all bacteria in one minute. Other investigations indicated that death rates for spores of *Bacillus* species was about 300 times greater with ozone than with chlorine.

Aquatic Organisms

Ozone is widely recommended to be used as a disinfectant for fish eggs in order to prevent the introduction of adhering disease agents. However, the application of ozone may have toxic side effects. Asbury and Coler (1980) determined the toxicity of dissolved ozone to eggs and larvae of a number of commercially important fish species. Results suggested that an alternative method to direct ozone application might be required because of the extremely high sensitivity of fish larvae to effective ozone concentrations. Eggs were less sensitive than the larvae.

Acevedo *et al.* (1982) found that exposure to low levels of ozone blocked the first cell division in sea urchin embryos (*Strongylocentrotus purpuratus*) if applied at 30 minutes post-insemination, but at 60 minutes post-insemination, cleavage was not blocked. Ozone specifically inhibits cell cleavage in embryo cells which do not grow during the early developmental period..

Ozone is presently being applied to seawater for shellfish depuration in France and Spain, for lobster culture recycling systems in Canada, and for fish culture in Europe. It is also being considered for use as an alternative to chlorine at coastal power plants for cooling-system biofouling control. However, a number of comparable studies show a similar toxicity of ozone-produced oxidants to early life-history stages of various fish species when compared to chlorine (Hall *et al.*, 1981). The results also indicate that eggs of striped bass are significantly more sensitive to ozonation in freshwater than in seawater.

Heath (1984) studied the decline in energy reserves of *Chlorella* cells upon exposure to ozone. Glucose-stimulated respiration, dependent upon ATP balance, is depressed to a greater degree than endogenous respiration in ozonated cells. Total ATP and glucose-6-phosphate levels also decrease, indicating that processes which both utilize and generate ATP are changed after ozone exposure and deplete the cell's energy reserves.

Hulle *et al.* (1982) determined the toxicity of ozone to *Nais communis*, an oligochaete worm species commonly found in water treatment plants in the sand and carbon filters. Ozone alone or in combination with chlorine efficiently stopped the worm at the first stage of the treatment process.

Hunter *et al.* (1981) investigated the effect of polar and artificial UV-B radiation on larval anchovy, but did not separate the effects of radiation from the oxidant by-product of the treatment.

Vertebrates and Man

Much knowledge has been gained through intensified study of ozone effects on lung physiology in a wide variety of vertebrates.

Examples of such studies are Abraham *et al.* (1984) looking at changes in airway permeability after ozone exposure, changes in antigen-induced bronchial spasm in relation to ozone concentration (Abraham *et al.*, 1983. Adams *et al.* (1981, 1983) identified ozone effects on pulmonary ventilation during endurance performance of athletes, noting altered exercise ventilatory patterns in trained athletes, suggesting endurance athletes may be more susceptible to the effects of a given ozone concentration than normal untrained adults. Alink *et al.* (1983) demonstrated that low ozone concentration (0.2 - 0.4 µg/L) had an acute toxic effect on human alveolar cells. STM studies revealed numerous ultrastructural changes, for example, loss of microvilli and dilation of microchondrial cristae. Radicals are probably involved in the mechanism of ozone toxicity.

In another study, Avol *et al.* (1984) could show that coexisting air pollutants don't enhance irritancy of ozone in moderate ambient oxidant pollution. Ozone itself, however, did produce respiratory irritation at 0.12 ppm when high ventilation rates are required during heavy exercise. Long-term exposure studies (Bery *et al.*, 1985) using the same ozone concentrations with rats revealed significant changes in the alveolar epithelium. Apparently, low ozone concentrations cause a chronic epithelial injury in the proximal alveolar region.

Bedi *et al.* (1985) could show that single initial short-term exposure to ozone for two hours at 0.45 ppm increased pulmonary function sensitivity leading to a detriment in the functional vital capacity, which however was soon repaired.

There is a difference in ozone sensitivity of the different airway zones, the nasal, tracheal, and bronchoalveolar. Bhalla *et al.* (1986) showed maximal ozone injury after 2 hours as reflected by permeability in the trachea and the bronchoalveolar regions, but injury persisted for less than 24 hours in the trachea, whereas it lasted longer than 24 hours in the bronchoalveolar zone. Apparently, ozone causes larger injury in the smaller airways and the alveolar zone than in the trachea. The 2-hour exposure of the previous study is of interest to aquatic workers because this time is similar to exposures when performing emergency service during equipment failure in ozonation plants. Similar exposures have also been shown for workers operating copy machines in poorly ventilated rooms (Gunter, 1981). Shepard *et al.* (1983) found that cigarette smokers showed less response and delayed response to 2 hour ozone exposure compared with non-smokers, suggesting the chronic effect of smoking may delay bronchial irritation by ozone.Lung function data have also been determined in 2 and 4 hour esposure when applied in single dose or in combination with other pollutant gases (Hackney *et al.*, 1975*a,b*).

Experiments performed by Gleiner *et al.* (1983) and Haak (1981) were designed to determine whether persons repeatedly exposed to low ozone (0.2 ppm) concentrations would demonstrate a diminished response as a result of adaptation or desensitization when subsequently exposed to a higher ozone concentration (0.5 ppm) but results showed that such adaptation does not persist.

Ozone is used in biochemical therapy of human blood. Washuettl *et al.* (1981) discuss effects on blood parameters in vitro and in vivo especially in cancer patients.

Safe concentration levels for human exposure to ozone were set by U.S. occupational health officials at relatively high levels in times past. About 1960 the American Conference of Government Industrial Hygenists (ACGIH) reduced the threshold limit for ozone from 1 ppm to 0.1 ppm and later in 1963 further reduced it from 0.1 to 0.05 ppm. The range of ozone exposures in welders of a maintenance department (Ohio, USA) was reported to reach 0.024 to 0.157 ppm (average 0.054 ppm). Because of the simultaneous exposure of welders to Cd and Ni, occasional over-exposure to ozone was assumed at the higher recorded levels (Albrecht and Boxer, 1982). Experiments with rats confirmed that a daily exposure to 0.3 ppm ozone for more than 3 hours would significantly impair the release of pulmonary surfactant (Shimura *et al.*, 1984).

Recent medical study results will soon lead to new adjustments of safety standards for short- and long-term exposure. It should be emphasized that ozone is a highly odiferous compound where a concentration of 0.1 ppm by weight in air is easily detected by the sense of smell. Therefore, ozone is in a sense self-policing. However, it is highly recommended that those working with ozone should be trained in its proper use and detection.

ATMOSPHERIC AND STRATOSPHERIC OZONE

In the fall of 1986 the 21 signatory States of the "Vienna Convention on Protection of the Ozone Layer" met for the first time, thereby attempting to tackle a potentially global environmental problem. Farman *et al.* (1985) were the first to report the rapid decrease of the total column amount of ozone in late winter and early spring over the Halley Bay Station in Antarctica (76 degrees S, 27 degrees W). These authors started their regular measurements in 1957. They attributed the decrease of ozone to the increase in stratospheric chlorine due to chlorofluorocarbon (CFC) release.

Manufacture and use of CFC's is a multi-billion dollar per year industry for companies such as the U.S.'s DuPont, York International, and Kaiser Chemical, Britain's Imperial Chemical Industries PLC, and Japan's Daikin Kogyo. In Canada and the U.S., non-essential use of CFC's for aerosol products such as hairspray has been banned since 1976. This caused an initial decline in demand for the CFC's involved, but the steadily growing use of the same CFC's for the production of foam rubber and foam insulating board and foam plastic packaging materials such as hamburger boxes and egg cartons, has resulted in annual demand levels of the CFC's rising again to 1976 peak levels. Industry is naturally hesitant to cease the production and use of CFC's without very convincing and conclusive evidence that CFC's are actually damaging the environment. Industry also points out that alternatives to chlorofluorocarbons take time to develop. These alternatives may be more expensive and/or not as efficient. In addition, the alternatives themselves may prove to be toxic or environmentally damaging, perhaps even more so than CFC's. Foam cushions, for example, can be produced using either CFC's or methylene chloride. The latter is more toxic to workers in the production plant.

Ozone depletion of the atmosphere, allowing higher levels of ultraviolet radiation to reach the surface of the earth and perhaps causing global warming, could have drastic ecological effects on the world. Predictions call for great increases in the number of cases of human skin cancers and resultant deaths from the UV radiation. The radiation would also affect plant and animal life. Global warming could also affect us drastically by melting the polar ice caps and raising world sea levels, *inter alia.*

Linking global ozone depletion to the release of CFC's is difficult and contoversial. While researchers have measured between 1978 and 1984 an annual mean atmospheric ozone erosion of 0.5%, this might be due to long-term atmospheric cycles beyond our present knowledge. Mechanisms to explain the formation of the antarctic ozone hole have been proposed by several authors, among them Callis and Natarajan (1986), Stolarski *et al.* (1986), Tung *et al.* (1986), McElroy *et al.* (1986) and Solomon *et al.* (1986). The mechanisms fall into two general categories, chemical or dynamic.

Stolarski *et al.* (1986) reported on global satellite measurements of the antarctic ozone decrease between 1979 and 1985, showing that the phenomenon is regional in extent. The ozone decrease begins during September as the sun rises, reaching a minimum in mid-October. They report a 40% decrease in the ozone minimum and a 20% decrease in the surrounding ozone maximum, based on their seven years (1979-1985) of October monthly mean values.

Callis and Natarajan (1986) measured stratospheric nitrogen-dioxide distribution by four different satellite experiments and found a mid-latitude increase of up to 75% during the 1979-1984 period. This increase in nitrogen dioxide provides in part an explanation for the dramatic springtime minima in the antarctic ozone. The authors also reported the lowest mid-latitude (North America, Europe, Asia) ozone levels since 1958 (the time of the last large solar maximum), and suggest that the ozone reductions are due in a large part to catalytic destruction by increasing nitrogen oxides through the two basic reactions:

$$
\begin{array}{ll}
(1) & NO + O_3 \rightarrow NO_2 + O_2 \\
(2) & NO_2 + O \rightarrow NO + O_2 \\
\hline
 & O_3 + O \rightarrow 2O_2
\end{array}
$$

Crutzen and Arnold (1986) have proposed a chemical mechanism for the formation of the ozone hole. It involves removal of gaseous odd nitrogen by ion- and/or aerosol-catalysed conversion of N_2O_5 and $ClONO_2$ to HNO_3 vapour, followed by heteromolecular HNO_3-H_2O condensation.

At present, a number of countries are undertaking major efforts to study the mechanisms in stratospheric and atmospheric ozone destruction and this has recently been reflected in a series of papers published in a special supplement issue of "Geophysical Research Letters" (Vol. 13, No. 12, November Supplement, 1986). In their overview paper, Schoeberl and Kreuger (1986) discuss difficulties that have been found with the various hypotheses attempting to explain the Antarctic ozone seasonal decline. They note that several papers show the decline in total ozone from 1979 to 1985 is correlated with a sizable decline (up to 18 degrees C) in stratospheric temperatures. Several papers have suggested that the ozone decline and the temperature decline can both be linked to small climatic shifts in the upper atmosphere. Redistribution processes between mid-latitude ozone and polar regions may also be a factor in seasonal changes of ozone in the Antarctic, complicating the issue of major causes of the "ozone hole."

In the meantime, negotiations in Geneva on controlling future emissions of CFC's are underway, aiming now to prevent further destruction of a band of ozone in the upper atmosphere that shields the earth from harmful ultraviolet radiation. The pros and cons discussed by industries and goverments indicate the lethargy of those institutional organizations that would have to take appropriate actions. Just what sort of gamble the world is willing to accept on the ozone layer may become clear in 1987 when participants to the Vienna Convention are supposed to arrive at a plan for CFC emission control (Crawford, 1986). The topic shall remain controversial until conclusive scientific evidence is able to provide guidance to industry and legislators. Shell (1987) provides a cogent overview of the situation.

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SUBJECT CATEGORY LIST

1.0 Atmospheric and Stratospheric Ozone.

- 1.1 General.
- 1.2 Urban pollution and local phenomena.
- 1.3 Large-scale and stratospheric phenomena.
- 1.4 Modelling of global distribution.

2.0 Preparation and Generation of Ozone.

- 2.1 General.
- 2.2 Ozonizer (electrical discharge).
  - 2.21 Using ambient air.
  - 2.22 Using oxygen enriched air.
  - 2.23 Using pure oxygen.
  - 2.24 Formation, removal, and/or oxidation of nitrogen oxides.
  - 2.25 Operational conditions of ozonizers.
- 2.3 Improvements in ozonizer technology.
  - 2.31 Pressure control gas feed, gas flow and dew control.
  - 2.32 Variable electrode discharge area.
  - 2.33 Special electrodes.
  - 2.34 Internal oxygen recycling.
  - 2.35 Special discharge output (microdischarge), including frequency control.
  - 2.36 Optimization of ozone output and ozone concentration control.
  - 2.37 Discharge coolants and cooling control.
  - 2.38 Miscellaneous.
- 2.4 Ozone generation by other processes.

3.0 Ozone Determination and Measurement.

- 3.2 Determination and measurement in air.
  - 3.21 Spectrophotometric, colorometric methods.
  - 3.22 Automatic reading devices.
  - 3.23 Intercalibration of methods.
  - 3.24 Electrochemical methods.
  - 3.25 In the presence of interfering gases and other substances.
  - 3.26 Miscellaneous.
- 3.3 Determination and measurement in water.
  - 3.31 Spectrophotometric, colorometric methods.
  - 3.32 Amperometric methods.
  - 3.33 Intercallibration of methods.
  - 3.34 Others.

4.0 Physical-Chemical Properties of Ozone.

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- 4.2 Chemical properties, structure and molecular weight.
- 4.3 Solubility in aqueous solutions.
- 4.4 Decomposition.
  - 4.41 In air.
  - 4.42 In aqueous solutions.
  - 4.43 In other media and on catalysts.
- 4.5 Interaction with elementary oxygen.
- 4.6 Interaction with other gases.

5.0 Chemical Reactions of Ozone.

- 5.1 Chemical reactions in general.
- 5.2 Ozonolysis and ozonolysis mechanism.
- 5.3 Ozonides.
  - 5.31 Ozonation products in general.
  - 5.32 Determination and properties of specific ozonides.
- 5.4 Organic compounds.
    - 5.401 Amines.
  - 5.41 In general.
  - 5.42 Criegee intermediates.
  - 5.43 Aliphatic compounds
    - 5.431 Olefins.
    - 5.432 Fatty acids, lipids, ketones, aldehydes.
  - 5.44 Organic acids (other than fatty acids).
    - 5.441 Amino acids.
  - 5.45 Esters.
  - 5.46 Alcohols and ethers.
  - 5.47 Proteins.
  - 5.48 Specific cyclic and semicyclic compounds.
    - 5.481 Aromatic compounds.
    - 5.482 Nucleic acids.
    - 5.483 Polycyclic compounds and polymers.
  - 5.49 Crude oils and products.
- 5.5 Elements and inorganic compounds.
  - 5.51 In general.
  - 5.52 Heavy metals.
  - 5.53 Nitrogen species.
  - 5.54 Gases in general
- 5.6 Chemical reactions in aqueous solutions.

6.0 Applications of Ozone.

- 6.1 Water treatment.
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    - 6.132 Polishing (taste).
    - 6.133 Sterilization (disinfection).
    - 6.134 Treatment of residual organics.
    - 6.135 Regional reports.
      - 6.1351 Europe.
      - 6.1352 North America.
      - 6.1353 Asia.
      - 6.1354 Other regions.
    - 6.136 Water treatment in breweries, the wine, must, and softdrink industries.
  - 6.14 Swimming pool water (private and public bath).
  - 6.15 Aquaculture (in general).
    - 6.151 Aquarium techniques.
    - 6.152 Quarantine stations.
    - 6.153 Recycling systems.
    - 6.154 Shellfish purification.
    - 6.155 Hatcheries.
    - 6.156 Treatment of toxins (i.e. red tides).
  - 6.16 Combined treatment (UV-ozone; ultrasound-ozone; activated charcoal-ozone).
  - 6.17 Combined action of ozone and chlorine.
  - 6.18 Comparison between ozone and chlorine treatment.
- 6.2 Municipal and industrial applications.
  - 6.21 Tertiary waste water treatment.
    - 6.211 Ozonation, settling and flocculation.
  - 6.22 Municipal and industrial waste water treatment.
    - 6.221 General.
    - 6.222 Municipal waste water treatment.
      - 6.2221 Pre-treatment.
      - 6.2222 Ammonia oxidation.
      - 6.2223 Nitrite oxidation.
      - 6.2224 Odor control in sewage.
    - 6.223 Industrial waste water treatment.
      - 6.2231 Phenols.
      - 6.2232 Cyanides.
      - 6.2233 Coal, coke, and gas works effluents.
      - 6.2234 Degradation of organics.

- 6.2235 COD-reduction.
- 6.2236 Crude oil and oil products (incl. refinery wastes).
- 6.2237 Pulp mill effluents.
- 6.2238 Off-gas control.
- 6.2239 Decoloration (incl. dye wastes).
- 6.224 Degradation of pesticides, herbicides, NTA, detergents, detoxification of heavy metals.
- 6.225 Treatment of seawater.
- 6.226 Reactions in combined treatment with activated carbon or charcoal.
- 6.23 Non-water treatment applications.
  - 6.231 Food treatment and food storage.
  - 6.232 Cosmetics.
  - 6.233 Medical science.
  - 6.234 Air treatment (sterilization).
  - 6.235 Treatment of materials.
    - 6.2351 In general
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- 7.5 Phytotoxicity (Plant injury).
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  - 7.54 Photochemistry (Photosynthesis).
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    - 7.622 *Escherichia coli*.
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  - 7.64 Protozoa.
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  - 7.72 Fish.
- 7.8 Insects.
- 7.9 Vertebrates and Man.
  - 7.91 General.
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  - 7.94 Lung physiology.
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    - 7.963 Medical studies.
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- 8.2 Resistant products.
- 8.3 Protection of materials. (Antiozonants).

9.0 Removal of Residual Ozone Gas.

10.0 Costs of Ozonation.

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- 11.2 Reviews.
- 11.3 General articles.

12.0 Ozone Patents.

- 12.1 Japan.
- 12.2 Germany.
- 12.3 USSR.
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- 12.5 Canada.
- 12.6 France.
- 12.7 UK.
- 12.8 Other Countries.

13.0 Miscellaneous Ozone Topics.

OZONE SUBJECT INDEX

1.0 OZONE IN THE ATMOSPHERE AND STRATOSPHERE.

1.1 General.

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3158. Nambu, Z., Yokoyama, E. 1982.
3311. Pagnotto, L.D., Epstein, S.S. 1969.
3348. Pauls, K.P., Thompson, J.E. 1982.
3467. Rakovski, S., Rakovski, K., Nenchev, L., Dobreva, R., Vyzharova, A. 1985.
3577. Rich, S., Ames, R., Zukel, J.W. 1974.
3578. Rich, S., Taylor, G.S. 1960.
3849. Seem, R.C., Cole, H., Lacasse, N.L. 1972.
3924. Siegel, S.M. 1962.
4393. Troquet, J., Cession-Fossion, A. 1964.
4500. Verweij, H., Van Steveninck, J. 1981.

7.4 Effects on cell structures and cell membranes (including permeability changes and chromosome damage).

302. Beckerson, D.W., Hofstra, G. 1980.
1135. Evans, L.S., Ting, I.P. 1973.
1202. Fetner, R.H. 1958.
1203. Fetner, R.H. 1959.
1204. Fetner, R.H. 1962.
1279. Freebairn, H.T. 1957.
1470. Goldstein, B.D., Lai, L.Y., Cuzzi-Spada, R. 1974.
1473. Goldstein, B.D., McDonagh, E.M. 1975.
1539. Grimes, H.D., Boss, W.F. 1982.
1581. Gukalina, T.V., Novosad, N.I., Kolodyaznaya, V.S. 1983.
1923. Holzman, R.S., Gardner, D.E., Coffin, D.L. 1968.
2375. Konev, S.V., Matus, V.K., Mel'nikova, A.M., Rudenok, A.N. 1982.
2382. Konyew, S.V., Kalyer, R.U., Matus, V.K., Yakubowski, S.M. 1982.
2433. Krivopishin, I.P. 1970.
2898. McFarlane, J.C., McNulty, I. 1968.
3169. Nasr, A.N.M. 1969.
3214. Nobel, P.S., Wang, C.-T. 1973.
3286. Ordin, L., Hall, M.A., Kindinger, J.I. 1969.
3287. Ordin, L., Skoe, B.P. 1965.
3310. Pace, D.M., Landolt, P.A., Alftonomos, B.T. 1969.
3368. Perkins, K.K., Boss, W.F. 1981.
3722. Sachsenmaier, W. 1960.
3723. Sachsenmaier, W., Siebs, W., Tan, T.-A. 1965.
3732. Sakaki, T., Ohnishi, J., Kondo, N., Yamada, M. 1985.
3908. Shinriki, N. 1983.
4033. Spotts, R.A., Lukezic, F.L., Hamilton, R.H. 1975.
4184. Swanson, E.S. 1971.
4251. Tchakhotine, S. 1937.
4761. Zelac, R.E., Cromroy, H.L., Bloch, W.E., Dunavant, B.G., Bevis, H.A. 1971.
4762. Zelac, R.E., Cromroy, H.L., Bloch, W.E., Dunavant, B.G., Bevis, H.A. 1971.
4771. Zhurkov, V.S., Pechennikova, E.V., Fel'dt, E.G., Garibyan, L.Kh., Tkhiem, T.K. 1979.

7.5 Phytotoxicity (plant injury).

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34. Agrawal, M., Nandi, P.K., Rao, D.N. 1982.
35. Agrawal, M., Nandi, P.K., Rao, D.N. 1985.
78. Amiro, B.D., Gillespie, T.J. 1985.
309. Bell, J.N.B., Cox, R.A. 1975.
338. Berry, C.R. 1970.
449. Brennan, E., Leone, I.A. 1972.
450. Brennan, E., Leone, I.A., Daines, R.H. 1964.
451. Brennan, E., Leone, I.A., Halisky, P.M. 1969.
586. Cantwell, A.M. 1968.
616. Cathey, H.M., Heggestad, H.E. 1982.
694. Clarke, B., Harkov, R., Brennan, E. 1980.
725. Comeau, G., Le Blanc, F. 1971.
733. Constantinidou, H.A., Kozlowski, T.T. 1979.
758. Craker, L.E. 1971.
853. Davis, D.D., Wood, F.A. 1972.
1014. Dugger, W.M., Koukol, J., Palmer, R.L. 1966.
1077. Elkiey, T., Ormrod, D.P. 1981.
1108. Ensing, J., Hofstra, G., Roy, R.C. 1985.
1313. Fujinuma, Y., Furukawa, A., Ushijima, T., Matsuoka, Y., Sato, A., Totsuka, I. 1984.
1701. Harris, M.J., Heath, R.L. 1981.
1775. Heagle, A.S., Heck, W.H., Body, D. 1971.
1790. Heck, W.W., Dunning, J.A., Hindawi, I.J. 1966.
1791. Heck, W.W., Taylor, O.C., Adams, R.M., Bingham, G., Miller, J.E., Preston, E.M., Weinstein, L.H. 1982.
1845. Hill, A.C., Pack, M.R., Treshow, M., Downs, R.J., Transtrum, L.G. 1960.
1846. Hill, A.C., Pack, M.R., Treshow, M., Downs, R.J., Transtrum, L.G. 1961.
1884. Hofstra, G., Beckerson, D.W. 1981.
1924. Homan, C. 1937.
2149. Jones, J.L. 1963.
2255. Kendrick, J.B., Darley, E.F., Middleton, J.T. 1962.
2353. Kohut, R.J., Davis, D.D., Merrill, W. 1976.
2565. Ledbetter, M.C., Zimmerman, P.W., Hitchcock, A.E. 1959.
2566. Lee, E.H., Bennett, J.H. 1981.
2606. Leone, I.A., Brennan, E. 1969.
2619. Letchworth, M.B., Blum, U. 1977.
2941. Menser, H.A., Heggestad, H.E. 1964.
2972. Middleton, J.T. 1959.
3216. Noland, T.L., Kozlowski, T.T. 1979.
3224. Nouchi, I., Mayumi, H., Yamazoe, F. 1984.
3278. Olszyk, D.M., Tingey, D.T. 1984.
3279. Olszyk, D.M., Tingey, D.T. 1984.
3281. Omasa, K., Hashimoto, Y., Aiga, I. 1981.
3296. Otto, H.W., Daines, R.H. 1969.
3434. Price, H.E. 1973.
3575. Rich, S. 1964.
3604. Richards, B.L., Taylor, O.C. 1965.
3725. Saio, K., Matsuyama, K., Enoki, M. 1978.
3846. Sechler, D., Davis, D.R. 1964.
4247. Taylor, G.E., Jr., Tingey, D.T., Ratsch, H.C. 1982.
4273. Thomas, M.D. 1951.
4277. Thompson, C.R., Kats, G., Hensel, E. 1972.
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4341. Tomlinson, H., Rich, S. 1967.
4390. Treshow, M. 1970.
4568. Wang, X. 1983.

7.51 Plant injury in general (including species other than listed below).

147. Ashmore, M., Bell, N., Rutter, J. 1985.
445. Brennan, E., Halisky, P.M. 1970.
447. Brennan, E., Leone, I.A. 1968.
453. Brewer, B.J., Reinert, R.A., Barker, K.R. 1980.
610. Castillo, F.J., Penel, C., Greppin, H. 1984.
612. Cathey, H.M., Heggestad, H.E. 1972.
613. Cathey, H.M., Heggestad, H.E. 1972.
614. Cathey, H.M., Heggestad, H.E. 1982.
762. Craker, L.E., Feder, W.A. 1972.
764. Craker, L.E., Starbuck, J.S. 1973.
829. Daines, R.H., Brennan, E., Leone, I.A. 1963.
830. Daines, R.H., Brennan, E., Leone, I.A., Lund, S. 1962.
852. Davis, D.D., Umbach, D.M., Coppolino, J.B. 1981.
939. Dijak, M., Ormrod, D.P. 1982.
1015. Dugger, W.M., Palmer, R.L. 1969.
1074. Elkiey, T., Ormrod, D.P. 1979.
1075. Elkiey, T., Ormrod, D.P. 1981.
1076. Elkiey, T., Ormrod, D.P. 1981.
1078. Elkiey, T., Ormrod, D.P. 1982.
1105. Engle, R.L., Gabelman, W.H., Romanowski, R.R. 1965.
1106. Engle, R.W., Gabelman, W.H. 1966.
1182. Feder, W.A., Campbell, F.J. 1968.
1371. Gentile, A.G., Feder, W.A., Young, R.E., Santner, Z. 1971.
1776. Heagle, A.S., Key, L.W. 1973.
1781. Heagle, A.S., Strickland, A. 1972.
1840. Hibben, C.R. 1969.
1868. Hodgson, R.H., Dusabek, K.E., Hoffer, B.L. 1974.
1873. Hoffman, G.J., Maas, E.V., Rawlins, S.L. 1975.
1937. Horsman, D.C., Nicholls, A.O., Calder, D.M. 1981.
1959. Howell, R.K., Thomas, C.A. 1972.
2101. James, R.L., Cobb, F.W., Jr., Parmeter, J.R., Jr. 1982.
2122. Jensen, K.F., Dochinger, L.S. 1974.

2143. Johnston, W.J., Haaland, R.L., Dickens, R. 1983.
2207. Karnosky, D.F., Steiner, K.C. 1981.
2340. Kobriger, J.M., Tibbitts, T.W., Brenner, M.L. 1984.
2419. Krause, C.R., Weidensaul, T.C. 1977.
2420. Krause, C.R., Weidensaul, T.C. 1978.
2421. Krause, C.R., Weidensaul, T.C. 1978.
2426. Kress, L.W., Skelly, J.M. 1982.
2529. Lantican, D.M., Cote, W.A., Skaar, C. 1965.
2581. Legassicke, B.C., Ormrod, D.P. 1981.
2607. Leone, I.A., Brennan, E. 1970.
2743. Mahoney, M.J., Skelly, J.M., Chevone, B.I., Moore, L.D. 1984.
2782. Manning, W.A., Feder, W.J., Perkins, I. 1972.
2785. Manning, W.J., Feder, W.A., Perkins, I. 1970.
2786. Manning, W.J., Feder, W.A., Perkins, I. 1972.
2882. McCool, P.M., Menge, J.A. 1983.
2883. McCool, P.M., Menge, J.A. 1984.
2885. McCool, P.M., Menge, J.A., Taylor, O.C. 1982.
3062. Mooi, J. 1981.
3063. Mooi, J. 1983.
3095. Mumford, R.A., Lipke, H., Laufer, D.A., Feder, W.A. 1972.
3276. Olszyk, D.M., Tibbitts, T.W. 1981.
3277. Olszyk, D.M., Tibbitts, T.W. 1981.
3280. Olszyk, D.M., Tingey, D.T. 1985.
3282. Omasa, K., Hashimoto, Y., Aiga, I. 1984.
3289. Ormrod, D.P., Adedipe, N.O., Hofstra, G. 1973.
3290. Ormrod, D.P., Adepipe, N.O., Hofstra, G. 1971.
3291. Ormrod, D.P., Tingey, D.T., Gumpertz, M.L., Olszyk, D.M. 1984.
3530. Reinert, R.A., Tingey, D.T., Carter, H.B. 1972.
3531. Reinert, R.A., Tingey, D.T., Carter, H.B. 1972.
3535. Reinhert, R.A., Gray, T.N. 1981.
3576. Rich, S. 1966.
3579. Rich, S., Tomlinson, H. 1968.
3616. Rist, D.L. 1983.
3617. Rist, D.L., Lorbeer, J.W. 1984.
3855. Semeniuk, P., Heggestad, H.E. 1981.
3892. Shew, B.B., Reinert, R.A., Barker, K.R. 1982.
3900. Shimizu, H., Motohashi, S., Iwaki, H., Furukawa, A., Totsuka, T. 1981.
4092. Stolzy, L.H., Taylor, O.C., Dugger, W.M., Mersereau, J.D. 1964.
4254. Temple, P.J., Taylor, O.C. 1985.
4303. Ting, I.P., Dugger, W.M. 1968.
4312. Tingey, D.T., Reinert, R.A., Dunning, J.A., Heck, W.W. 1973.
4327. Todd, G.W., Arnold, W.N. 1961.
4376. Townsend, A.M., Dochinger, L.S. 1974.
4655. Wilton, A.C., Murray, J.J., Heggestad, M.E., Juska, F.V. 1972.
4671. Wukasch, R.T., Hofstra, G. 1977.
4672. Wukasch, R.T., Hofstra, G. 1977.

7.52 Species and species groups.

133. Applegate, H.G., Durrant L.C. 1969.
148. Ashmore, M.R., Onal, M. 1984.
697. Clayberg, C.D. 1971.
820. Czuba, M., Ormrod, D.P. 1981.
3872. Shaulis, N.J., Kender, W.J., Pratt, C., Sinclair, W.A. 1972.

7.521 Unicellular algae.

347. Betzer, N., Argaman, Y., Kott, Y. 1980.
1278. Frederick, P.E., Heath, R.L. 1975.
1432. Ginocchio, J.C. 1981.
1433. Ginocchio, J.C. 1981.
1434. Ginocchio, J.C. 1981.
1785. Heath, R.L. 1984.
1786. Heath, R.L., Frederick, P.E., Chimiklis, P.E. 1982.
4185. Swanson, E.S., Toivio-Kinnucan, M., Heath, R., Cunningham, W.P. 1982.
4347. Toner, R.C., Brooks, B. 1975.
4498. Verkroost, M. 1974.
4520. Vokk, R.A., Sukhareva-Nemakova, N.N. 1980.

7.522 Overall crop losses.

1021. Dunning, J.A., Heck, W.W., Tingey, D.T. 1974.
1778. Heagle, A.S., Letchworth, M.B., Mitchell, C.A. 1983.
1779. Heagle, A.S., Letchworth, M.B., Mitchell, C.A. 1983.
1788. Heck, W.W., Adams, R.M., Cure, W.W., Heagle, A.S., Heggestad, H.E., Kohut, R.J., Kress, L.W., Rawlings, J.O., Taylor, O.C. 1983.
1960. Howitt, R.E., Gossard, T.W., Adams, R.M. 1984.
3488. Rawlings, J.O., Cure, W.W. 1985.

7.523 Conifers.

256. Barnes, R.L. 1972.
257. Barnes, R.L. 1972.
323. Benoit, L.F., Skelly, J.M., Moore, L.D., Dochinger, L.S. 1982.
339. Berry, C.R. 1971.
340. Berry, C.R., Ripperton, L.A. 1962.
349. Bialobok, S., Karolewski, P., Oleksyn, J. 1980.
411. Boone, G.C. 1980.
414. Botkin, D.B., Smith, W.H., Carlson, R.W. 1971.
415. Botkin, D.B., Smith, W.H., Carlson, R.W., Smith, T.L. 1972.
446. Brennan, E., Leone, I., Harkov, R., Rhoads, A. 1981.
745. Costonis, A.C. 1970.
746. Costonis, A.C. 1971.
747. Costonis, A.C. 1973.
748. Costonis, A.C., Sinclair, W.A. 1969.
749. Costonis, A.C., Sinclair, W.A. 1969.
756. Coyne, P.I., Bingham, G.E. 1981.
854. Davis, D.D., Wood, F.A. 1973.
855. Davis, D.D., Wood, F.A. 1973.
965. Dochinger, L.S., Bender, F.W., Fox, F.L., Heck, W.W. 1970.
1132. Evans, L.S. Miller, P.R. 1975.
1133. Evans, L.S., Miller, P.R. 1972.
1134. Evans, L.S., Miller, P.R. 1972.
1952. Houston, D.B. 1974.
2428. Kress, L.W., Skelly, J.M., Hinkelmann, K.H. 1982.
2429. Kress, L.W., Skelly, J.M., Hinkelmann, K.H. 1982.
2658. Linzon, S.N. 1967.
2990. Miller, P.R., Parmeter, J.R., Flick, B.H., Martinez, C.W. 1969.
2991. Miller, P.R., Parmeter, J.R., Taylor, J.R., Cardiff, E.A. 1963.
3105. Murdoch, C.W., Campana, R.J. 1980.
3605. Richards, B.L., Taylor, O.C., Edmunds, G.F. 1968.
3952. Sinclair, W.A., Costronis, A.C. 1967.
4314. Tingey, D.T., Wilhour, R.G., Stadley, C. 1976.
4392. Trimble, J.L., Skelly, J.M., Tolin, S.A., Orcutt, D.M. 1982.

7.524 Beans.

22. Adedipe, N.O., Hofstra, G., Ormrod, D.P. 1972.
78. Amiro, B.D., Gillespie, T.J. 1985.
79. Amiro, B.D., Gillespie, T.J., Thurtell, G.W. 1984.
82. Amthor, J.S., Fong, F. 1981.
547. Butler, L.K., Tibbetts, T.W. 1979.
763. Craker, L.E., Starbuck, J.S. 1972.
814. Curtis, C.R., Howell, R.K. 1971.
847. Dass, H.C., Weaver, G.M. 1972.
1131. Evans, L.S. 1973.
1136. Evans, L.S., Ting, I.P. 1974.
1235. Fletcher, R.A., Adedipe, N.O., Ormrod, D.P. 1972.
1796. Heggestad, H.E., Heagle, A.S., Bennett, J.H., Koch, E.J. 1980.
1883. Hofstra, G., Ali, A., Wukasch, R.T., Fletcher, R.A. 1981.
1968. Hucl, P., Beversdorf, W.D. 1982.
1969. Hucl, P., Beversdorf, W.D., McKersie, B.D. 1982.
2067. Ito, O, Mitsumori, F., Totsuka, T. 1984.
2068. Ito, O., Mitsumori, F., Totsuka, T. 1985.
2069. Ito, O., Okano, K., Kuroiwa, M., Totsuka, T. 1984.
2070. Ito, O., Okano, K., Kuroiwa, M., Totsuka, T. 1985.
2071. Ito, O., Okano, K., Totsuka, T. 1984.
2567. Lee, E.H., Bennett, J.H. 1982.
2906. McKersie, B.D., Hucl, P., Beversdorf, W.D. 1982.
2910. McLaughlin, S.B., McConathy, R.K. 1983.
2958. Meredith, F.I., Thomas, C.A., Heggestad, H.E. 1986.
3123. Musselman, R.C., Huerta, A.J., McCool, P.M., Oshima, R.J. 1986.
3266. Okano, K., Ito, O., Takeba, G., Shimizu, A., Totsuka, T. 1984.
3267. Okano, K., Ito, O., Takeba, G., Shimizu, A., Totsuka, T. 1984.
3268. Okano, K., Ito, O., Takeba, G., Shimizu, A., Totsuka, T. 1984.
3269. Okano, K., Ito, O., Takeba, G., Shimizu, A., Totsuka, T. 1985.
3423. Prasad, K., Weigle, J.L., Sherwood, C.H. 1970.
3580. Rich, S., Tomlinson, H. 1968.
3581. Rich, S., Waggoner, P.E., Tomlinson, H. 1970.
3701. Rufner, R., Witham, F.H., Cole, H., Jr. 1975.
3705. Runeckles, V.C., Resh, H.M. 1975.
3706. Runeckles, V.C., Rosen, P.M. 1974.
3764. Satoh, S., Fujiwara, T. 1983.
4034. Spotts, R.A., Lukezie, F.L., Lacasse, N.L. 1975.
4053. Stan, H.-J., Schicker, S. 1982.
4054. Stan, H.-J., Schicker, S., Kassner, H. 1981.
4285. Thomson, W.W., Dugger, W.M., Palmer, R.L. 1966.
4329. Toivonen, P.M.A., Hofstra, G., Wukasch, R.T. 1982.

4344. Tomlinson, H., Rich, S. 1970.
4345. Tomlinson, H., Rich, S. 1971.
4348. Tonneijek, A.E.G. 1983.
4588. Weaver, G.H., Jackson, G.O. 1968.

7.5241 Soybeans.

454. Brewer, P.F., Heagle, A.S. 1983.
654. Chevone, B.I., Yang, Y.S. 1985.
1286. Frey, J.E. 1972.
1777. Heagle, A.S., Letchworth, M.B. 1982.
1795. Heggestad, H.E., Gish, T.J., Lee, E.H., Bennett, J.H., Douglass, L.W. 1985.
1958. Howell, R.K., Kremer, D.F. 1972.
2142. Johnston, J.W., Jr., Heagle, A.S. 1982.
3221. Norby, R.J., Luxmoore, R.J. 1983.
3438. Prokipcak, B., Ormrod, D.P. 1986.
3516. Reich, P.B., Amundson, R.G., Sawicki, D., Carlsson, L.I., Lassoie, J.P. 1981.
3521. Reich, P.B., Schoettle, A.W., Amundson, R.G. 1985.
4306. Tingey, D.T. 1972.
4307. Tingey, D.T., Fites, R.C., Wickliff, C. 1973.
4311. Tingey, D.T., Reinert, R.A., Carter, H.B. 1972.

7.5242 Pinto beans.

626. Chang, C.W. 1971.
627. Chang, C.W. 1971.
628. Chang, C.W. 1972.
1016. Dugger, W.M., Taylor, O.C., Cardiff, E., Thompson, C.R. 1962.
1104. Engle, R.L., Gabelman, W.H. 1967.
1252. Fong, F., Heath, R.L. 1981.
1789. Heck, W.W., Dunning, J.A. 1967.
1872. Hoffman, G.J., Maas, E.V., Rawlins, S.L. 1973.
2730. MacKnight, M. 1968.
2783. Manning, W.J., Feder, W.A., Papia, P.M., Perkins, I. 1971.
2982. Miller, C.A., Davis, D.D. 1981
3355. Pell, E.J., Brennan, E. 1973.
3358. Pellissier, M., Lacasse, N.L., Cole, H. 1972.
3359. Pellissier, M., Lacasse, N.L., Ercegovich, C.D., Cole, H. 1972.
3849. Seem, R.C., Cole, H., Lacasse, N.L. 1972.
4313. Tingey, D.T., Thutt, G.L., Gumpertz, M.L., Hogsett, W.E. 1982.
4377. Townsend, B.B. 1965.
4596. Weidensaul, T.C. 1982.

7.525 Turnip.

1774. Heagle, A.S., Cure, W.W., Rawlings, R.O. 1985.

7.526 Tobacco.

21. Adedipe, N.O., Fletcher, R.A., Ormrod, D.P. 1973.
360. Bisessar, S., Palmer, K.T. 1984.
830. Daines, R.H., Brennan, E., Leone, I.A., Lund, S. 1962.
860. De Vos, N.E., Hill, R.R., Jr., Pell, E.J., Cole, R.H. 1982.
861. Dean, C.E., Davis, D.R. 1967.
1181. Feder, W.A. 1968.
1359. Gardner, W.S. 1973.
1789. Heck, W.W., Dunning, J.A. 1967.
1797. Heggestad, H.E., Middleton, J.T. 1959.
2574. Lee, T.T. 1965.
2575. Lee, T.T. 1966.
2576. Lee, T.T. 1967.
2577. Lee, T.T. 1968.
2608. Leone, I.A., Brennan, E., Daines, R.H. 1966.
2718. Macdowall, F.D.H. 1965.
2719. Macdowall, F.D.H. 1965.
2721. Macdowall, F.D.H., Cole, A.F.W. 1971.
2722. Macdowall, F.D.H., Cole, A.F.W., Katz, M., Mukammel, E.I. 1962.
2723. Macdowall, F.D.H., Ludwig, A.R. 1962.
2724. Macdowall, F.D.H., Mukammal, E.I., Cole, A.F.W. 1964.
2725. Macdowall, F.D.H., Vickery, L.S., Runeckles, V.C., Patrick, Z.A. 1963.
2937. Menser, H.A. 1963.
2938. Menser, H.A. 1964.
2939. Menser, H.A. 1966.
2940. Menser, H.A. 1969.
2942. Menser, H.A., Heggestad, H.E. 1966.
2943. Menser, H.A., Heggestad, H.E., Grosso, J.J. 1966.
2944. Menser, H.A., Heggestad, H.E., Street, O.E. 1963.
2945. Menser, H.A., Heggestad, H.E., Street, O.E., Jeffrey, R.N. 1963.
2946. Menser, H.A., Hodges, G.H. 1967.
2947. Menser, H.A., Hodges, G.H. 1968.
2948. Menser, H.A., Hodges, G.H. 1969.
2949. Menser, H.A., Hodges, G.H. 1970.
2950. Menser, H.A., Sorokin, T., Engelhaupt, M.E. 1965.
3091. Mukammel, E.I. 1965.
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